

# A benign, low Z electron capture agent for negative ion TPCs

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## Abstract

We have identified nitromethane ( $\text{CH}_3\text{NO}_2$ ) as an effective electron capture agent for negative ion TPCs (NITPCs). We present drift velocity and longitudinal diffusion measurements for negative ion gas mixtures using nitromethane as the capture agent. Not only is nitromethane substantially more benign than the only other identified capture agent,  $\text{CS}_2$ , but its low atomic number will enable the use of the NITPC as a photoelectric X-ray polarimeter in the 1-10 keV band.

*Key words:* , Dark Matter, negative ion, X-ray polarimeter, diffusion, ion mobility

*PACS:* 29.40.Cs, 51.20.+d, 51.50.+v, 52.70.La, 95.35.+d, 95.55.Qf

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## 1 Introduction

The negative ion time projection chamber (NITPC) achieves high spatial resolution by transporting charge in the form of negative ions, rather than electrons, thereby reducing diffusion to the thermal limit in both the transverse and longitudinal drift directions[1]. This provides the highest 3-D space-point resolution attainable for long drift distances, particularly where operation at low pressures is required to make very low energy tracks long enough to measure (for example, dark matter WIMP recoil atoms). In relatively low rate

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environments, the NITPC also has the advantage that the ion drift velocities are about two orders-of-magnitude less than those of electrons. This allows the use of low-bandwidth electronics that results in lower noise and power consumption.

The key to reducing diffusion to the thermodynamic lower limit (Equation 1 below) is to prevent net energy transfer from the drift field to the drifting primary ionization[2]. Such “heating” of the drifting charge occurs when electrons are drifted at reduced fields  $E/p > \sim 10\text{-}50 \text{ V/m}\cdot\text{Torr}$ . Heating is reflected in an increase of the parameter  $\epsilon$  in Equation 1 to values higher than  $\frac{3}{2}kT$ , resulting in increased diffusion. For electron drift, gas additives with large integrated cross sections for inelastic scattering of electrons, such as hydrocarbons and  $\text{CO}_2$ , provide channels for dissipation and are known [2,3] to prevent heating of drifting electrons up to moderate values of the reduced field ( $\sim 50 \text{ V/m}\cdot\text{Torr}$ ).

In contrast, ions drifting in even a high field are very efficiently thermally coupled to the room-temperature gas, since the ion mass is comparable to that of the gas molecules. Then, even elastic scattering produces substantial energy transfer from the drifting ion to the bulk gas. Ion diffusion has been measured in a number of  $\text{CS}_2$  mixtures suitable for TPC operation[1,4,5] and was found to follow the “thermal, low field” limiting behavior[2]:

$$\sigma_D = \sqrt{\frac{4\epsilon L}{3eE}} \quad (1)$$

Here  $\sigma_D$  is the rms diffusion spread for carriers with charge  $e$  after drifting through a distance  $L$  in electric field  $E$  with average (thermal) energy  $\epsilon$ . The parameter  $\epsilon$  (the average thermal energy of a drifting charge carrier) remained approximately equal to the room-temperature value, up to reduced drift fields as high as  $\sim 2.5 \times 10^3 \text{ V/m}\cdot\text{Torr}$ .

Many chemical vapors at STP will capture thermal electrons to form negative ions[6]. However, to maintain good track resolution the agent must have a capture mean free path of a hundred microns or less at a relatively low partial pressure. Since gas gain is desired at the TPC endcaps, the agent must also relinquish its electrons in the high, strongly inhomogeneous field of the gain structures, allowing Townsend avalanches to form. In all previous work,  $\text{CS}_2$  has been used as the only capture agent fulfilling these requirements reasonably well.

While the NITPC has been successfully implemented with  $\text{CS}_2$  as the capture agent in dark matter observatories[7–9], the utility of the NITPC would be expanded by the identification of new capture agents.  $\text{CS}_2$  has a combination of low flash point ( $-30^\circ \text{C}$ ), high vapor pressure (400 Torr) and low explosive mix-

63 ture limit in air (1.3%) which require great care in handling the material[10].  
 64 Nitromethane[11] has a flash point far above room temperature and a vapor  
 65 pressure less than a tenth that of CS<sub>2</sub>. The comparative health effects of the  
 66 two materials are indicated by the respective ACGIH Threshold Limit Values  
 67 of 1 ppm for CS<sub>2</sub> vs. 20 ppm for nitromethane. CS<sub>2</sub> also may form sulfide  
 68 deposits under conditions of high rate or discharge. More importantly, new  
 69 applications for the NITPC would be enabled by capture agents with different  
 70 functional properties. For example, a single-ion counting NITPC with high  
 71 energy resolution for double beta decay searches has been proposed[12] that  
 72 would benefit from a capture agent with higher ion mobility than CS<sub>2</sub>. An-  
 73 other example is photoelectric X-ray polarimetry below 10 keV with a NITPC,  
 74 which requires capture agents with lower atomic number (see below).

75 Photoelectric polarimeters derive their information by determining the photo-  
 76 electron emission direction, which is strongly correlated with the electric field  
 77 vector of the incident photon. The most sensitive photoelectric polarime-  
 78 ters image the tracks of the photoelectrons with pixels small compared to the  
 79 photoelectron track length and reconstruct the emission direction on an event-  
 80 by-event basis. Such polarimeters have been realized with optical avalanche  
 81 chambers[13,14], micropattern gas detectors with direct pixel readout[15–17],  
 82 and a micropattern TPC[18].

83 X-ray polarimetry below 10 keV with a NITPC would benefit from low atomic  
 84 number capture agents for several reasons. First, the correlation between  
 85 the photoelectron emission direction and photon electric field is strongest for  
 86 emission from s-orbitals[19], so it is preferable to use gases whose K-shell  
 87 energies are well below the X-ray energy. Second, the photoelectron is emitted  
 88 with a kinetic energy that is the difference between the binding energy and the  
 89 X-ray energy so that lower Z gases result in photoelectrons with larger kinetic  
 90 energies and longer track lengths. Finally, lower Z gases have less multiple  
 91 Coulomb scattering, which results in straighter tracks and a better correlation  
 92 between the photoelectron emission direction and photon electric field.

93 Therefore, motivated by the requirements of low energy X-ray polarimetry and  
 94 guided by the chemical literature on electron capture, we have explored a num-  
 95 ber of compounds as alternative NITPC capture agents. We have identified  
 96 nitromethane (CH<sub>3</sub>NO<sub>2</sub>) as a suitable capture agent for X-ray polarimetry  
 97 which is also relatively benign. We present measurements of drift velocity,  
 98 gas gain and longitudinal diffusion in gases with nitromethane as the capture  
 99 agent.

## 100 2 Capture agent screening apparatus

101 Capture agents were screened for negative ion drift velocity, gas gain and  
102 longitudinal diffusion at low to moderate fields using a single-wire proportional  
103 counter attached to a homogeneous-field drift region. The apparatus was  
104 enclosed in a stainless steel bell jar with a simple gas manifold. Drift fields  
105 up to  $4.0 \times 10^4$  V/m were used with this apparatus at reduced pressure.  
106 Photoelectrons were generated at an Sn photocathode attached to the drift-  
107 cathode. The photocathode was arranged so that it could be cleaned between  
108 runs using a glow discharge in pure argon. This was essential for maintaining  
109 the photoelectron yield.

110 UV light flashes from an EG&G Flash-Pak[20] were admitted into the bell jar  
111 through a quartz window, passed through a hole in the proportional counter  
112 wall, and struck the photocathode producing photoelectrons. The standard  
113 internal capacitors of the Flash-Pak were augmented with additional HV ca-  
114 pacitors to give a stored energy of about 0.2 Joule per pulse. The Flash-Pak  
115 was triggered by an external pulser, from which a time-zero signal was also de-  
116 rived. The proportional wire signal was read out through an ORTEC 142PC  
117 preamp and an ORTEC 579 shaping amplifier. Drift times to the proportional  
118 wire and wire-signal time widths were measured with a digital oscilloscope.

119 Drift velocities and gas gain at moderate to high fields ( $1.0 \times 10^5$  -  $4.0 \times 10^5$   
120 V/m) were measured in various gas mixtures with a micropattern gas detector  
121 (MPGD) having the same electrode structure as the gas electron multiplier[21],  
122 but assembled from two etched stainless steel meshes[18] separated by a 100  
123  $\mu\text{m}$  thick teflon spacer. A drift electrode, also of stainless steel, was placed  
124 10 mm above the MPGD cathode. The MPGD was operated at a gain of  
125 about 3000. The MPGD cathode was instrumented with a charge-sensitive  
126 preamplifier followed by a bipolar shaping amplifier with a six microsecond  
127 shaping time constant.

128 The MPGD cathode and drift electrode were simultaneously illuminated with  
129 a xenon flashlamp (Perkin-Elmer PAX-10), which produced photoelectrons  
130 from both surfaces. Photoelectrons from the cathode produced a prompt  
131 pulse, while the photoelectrons from the drift electrode produced negative ions  
132 and gave a pulse delayed by the drift time across the 10mm drift distance. The  
133 drift time was measured by averaging 100 pulses on a digital oscilloscope and  
134 taking the difference between the zero-crossing times of the prompt and the  
135 delayed pulses.

### 136 3 Results and Discussion

137 Nitromethane is known to have a large capture cross section for thermal elec-  
138 trons[6]. However, at moderate to high drift fields the pure near-saturated  
139 vapor at 20 Torr produces both electron-drift and negative-ion-drift signals.  
140 This is understandable since a drift field of  $5.0 \times 10^4$  V/m at 20 Torr is al-  
141 ready a reduced field of 2500 V/m·Torr, easily high enough to raise the energy  
142 of drifting electrons out of the thermal range before they could be captured.  
143 CO<sub>2</sub> was therefore added to the nitromethane vapor. CO<sub>2</sub> is known to be very  
144 effective in thermalizing drifting electrons, due to its large inelastic scatter-  
145 ing cross sections[3]. Mixtures of 20 Torr nitromethane with 50 Torr or more  
146 of CO<sub>2</sub> were found to have satisfactory characteristics as negative ion drift  
147 mixtures.

148 Drift velocity and diffusion results for the nitromethane:CO<sub>2</sub> 20:50 mixture  
149 are shown in Figures 1 and 2. The drift velocity rises linearly with drift field,  
150 with mobility  $(4.27 \pm .03) \times 10^{-4}$  m<sup>2</sup>/V·sec and a small positive intercept  
151  $(1.44 \pm .08$  m/s). The diffusion is studied by plotting the square of the pulse  
152 width in time as a function of  $1/(v_d^2 E_d)$ . The pulse width is expected to be the  
153 quadrature sum of the amplifier shaping time (10  $\mu$ sec) and the broadening  
154 due to diffusion. If the diffusion is governed by Equation 1, this plot will  
155 be linear in the region where diffusion dominates (essentially the entire plot),  
156 with slope. Fitting Figure 2 to a straight line gives  $T = 313 \pm 25$  K.

157 Drift velocities for higher pressure gas mixtures of nitromethane-CO<sub>2</sub>-neon,  
158 nitromethane-CO<sub>2</sub>-argon and nitromethane-CH<sub>4</sub> were measured up to drift  
159 fields of  $4.0 \times 10^5$  V/m. Results are shown in Figure 3. The linear dependence  
160 of drift velocity on drift field persists up to these very high fields. Results of  
161 linear fits for the mixtures studied are shown in Table 1. Shown in Figure  
162 4 are gas gain curves for higher pressure mixtures of CO<sub>2</sub>-neon, and neon-  
163 CO<sub>2</sub>-nitromethane. Gains as high as several thousands were obtained in all  
164 mixtures demonstrating the ability of nitromethane as a feasible, productive  
165 detector gas.

### 166 4 Conclusion

167 A negative ion drift mixture of nitromethane with CO<sub>2</sub> is found to have linear  
168 drift velocity vs. field and to exhibit thermal-limit longitudinal diffusion up to  
169 drift fields as high as  $2.8 \times 10^4$  V/m. Higher pressure mixtures with argon,  
170 neon, CH<sub>4</sub> and CO<sub>2</sub> also show linear drift velocity up to fields as high as  $4.0$   
171  $\times 10^5$  V/m. All mixtures exhibit stable operation as NITPCs.

172 The introduction of nitromethane as an electron capture agent will enable the  
173 use of the NITPC as an X-ray polarimeter in the 1-10 keV band. The low drift  
174 velocity of the NITPC greatly eases the difficulty of calibrating drift velocity  
175 and substantially reduces power consumption, which is particularly important  
176 for satellite-borne astronomical instruments. The low diffusion of the NITPC  
177 will also enable large-volume photoelectric polarimeters that could be used,  
178 for example, as wide field-of-view instruments for unpredictable astronomical  
179 transients, such as gamma-ray bursts, or with rotation modulation collimators  
180 for high-resolution imaging polarimetry of solar flares[22].

## 181 **Acknowledgements**

182 This work was performed, in part, under NASA contract **NNG07EJ03C**.

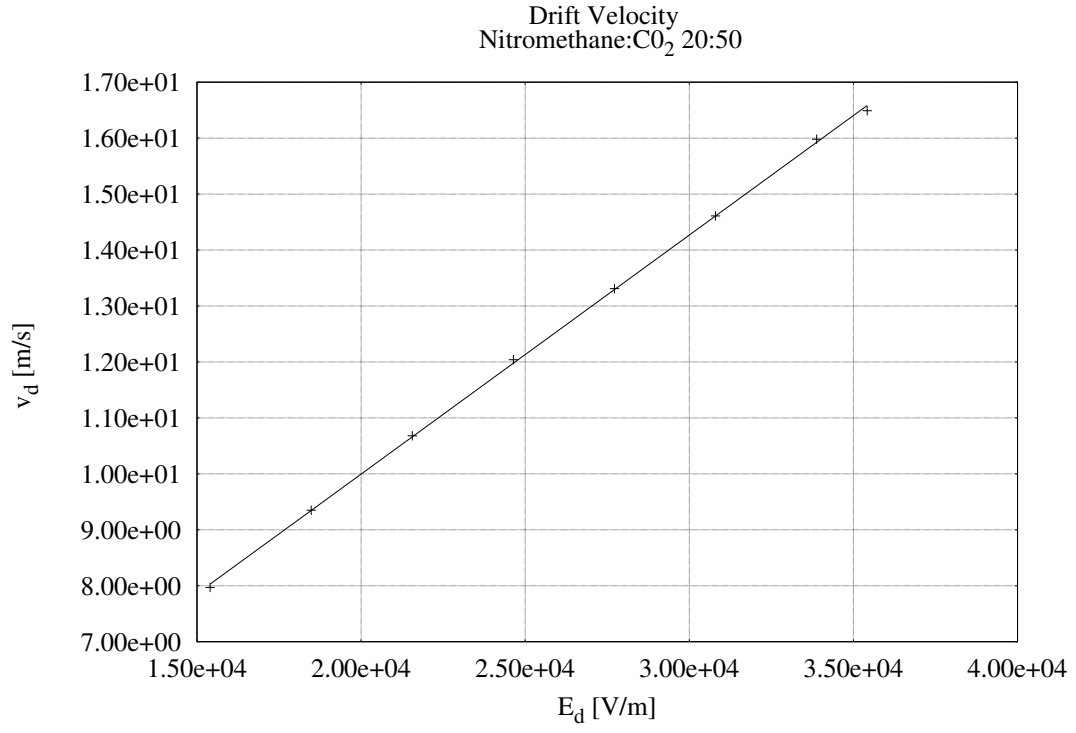


Fig. 1. Drift velocity  $v_d$  vs. drift field  $E_d$  for Nitromethane:CO<sub>2</sub> 20:50

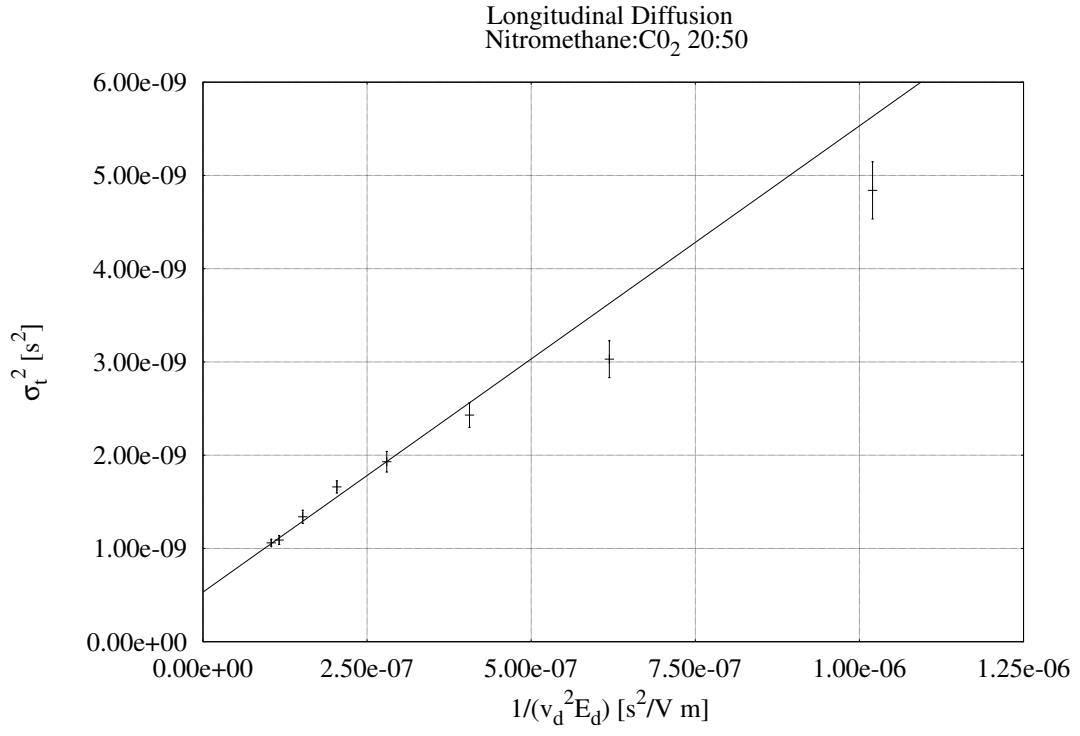


Fig. 2. Longitudinal diffusion data for Nitromethane:CO<sub>2</sub> 20:50, plotted as the square of the pulse time width vs.  $1/v_d^2 E_d$  for 80 mm drift.



Mixture	CO <sub>2</sub> Torr	Neon Torr	Argon Torr	Methane Torr	Mobility [m <sup>2</sup> /V·s]
A	510	170	–	–	$8.43 \pm .01 \times 10^{-5}$
B	170	-	510	–	$9.69 \pm .01 \times 10^{-5}$
C	340	340	–	–	$10.5 \pm .01 \times 10^{-5}$
D	170	510	–	–	$13.1 \pm .02 \times 10^{-5}$
E	–	–	–	380	$21.0 \pm .07 \times 10^{-5}$

Table 1

Fitted slopes for ion drift in mixtures of 20 Torr Nitromethane with CO<sub>2</sub>, Neon, Argon and Methane.

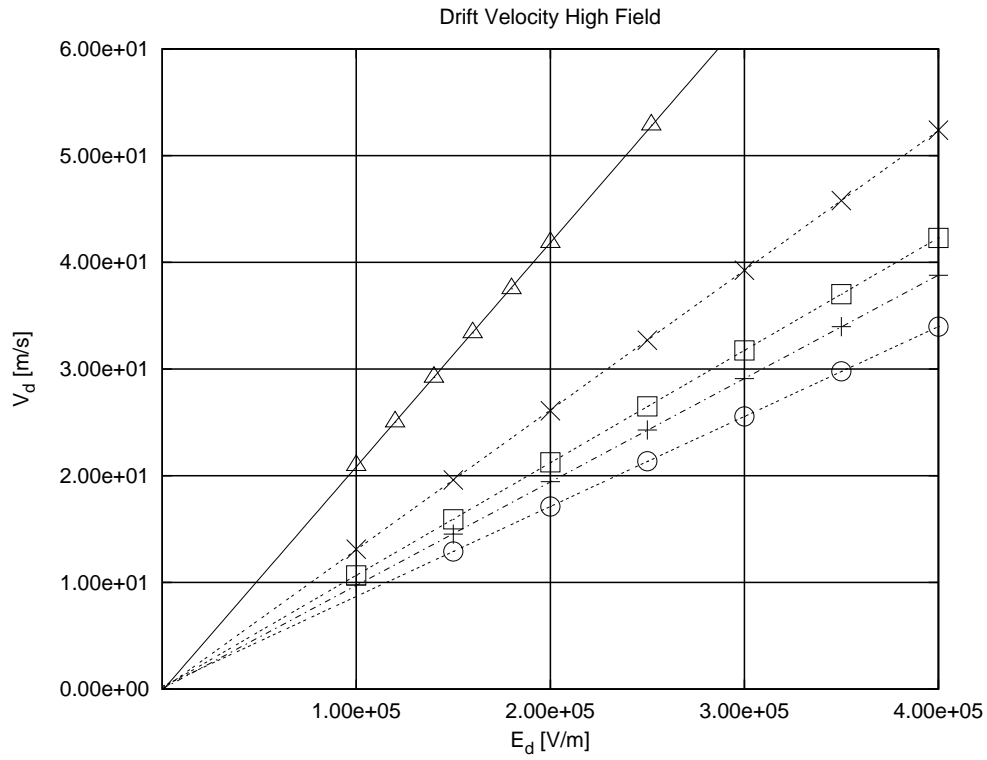


Fig. 3. Drift velocity at high fields in mixtures of Nitromethane and CO<sub>2</sub> and/or Neon, Methane mixtures A-E described in Table 1. Circles: A, Plusses: B, Squares: C, Crosses: D, Triangles: E

Mixture	CO <sub>2</sub> Torr	Neon Torr	Nitromethane
A	210	490	-
B	170	510	20
C	340	340	20
D	510	170	20

Table 2

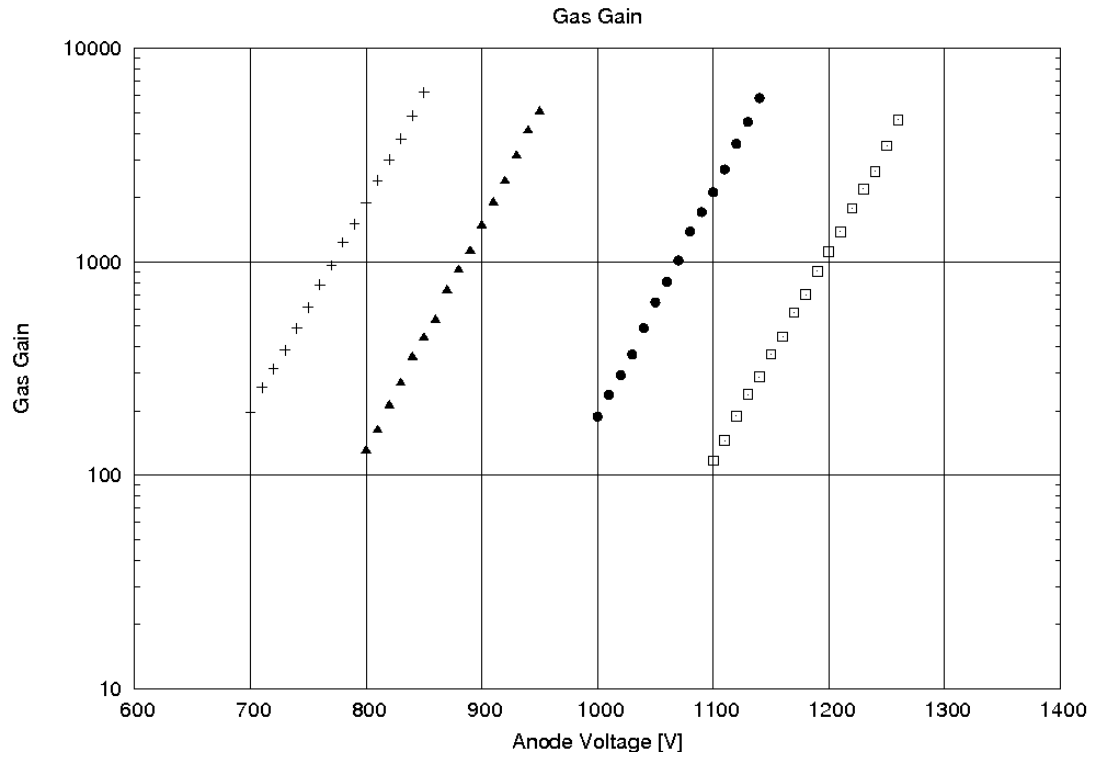


Fig. 4. Gas Gain in mixtures containing Neon and CO and/or Nitromethane A-D described in Table 2. Plusses: A, Triangles: B, Circles: C, Squares: D

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